

# Polarization Properties of Single Quantum Dots in Nanowires

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We study the absorption and emission polarization of single semiconductor quantum dots in semiconductor nanowires. We show that the polarization of light absorbed or emitted by a nanowire quantum dot strongly depends on the orientation of the nanowire with respect to the directions along which light is incident or emitted. Light is preferentially linearly polarized when directed perpendicular to the nanowire elongation. In contrast, the degree of linear polarization is low for light directed along the nanowire. This result is vital for photonic applications based on intrinsic properties of quantum dots, such as generation of entangled photons. As an example, we demonstrate optical access to the spin states of a single nanowire quantum dot.

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Semiconductor quantum dots (QDs) are sources of single [1, 2] and entangled photons [3, 4, 5], single electrons [6], and are naturally integrated with modern semiconductor electronics. Incorporating them in semiconducting nanowires (NWs) [7, 8, 9, 10, 11] brings additional unique features such as natural alignment of vertically stacked QDs [12] and an inherent one-dimensional channel for charge carriers. Furthermore, the unprecedented material and design freedom makes them very attractive for novel opto-electronic devices [13, 14, 15, 16, 17, 18] and quantum information science in general [19, 20, 21, 22]. However, access to intrinsic spin and polarization properties of a QD in a NW has never been demonstrated, most likely due to an insufficient quality of the NW QDs. Moreover, the NW geometry strongly affects the polarization of photons emitted or absorbed by a NW QD, and thus becomes the main obstacle for applications based on intrinsic spin or polarization properties of QDs such as an electron spin memory [23] or generation of entangled photons [3]. Indeed, it has been shown that luminescence of pure NWs is highly linearly polarized and the polarization direction is parallel to the NW elongation [13, 17, 24].

In this Letter we demonstrate that by directing light along the NW elongation we can access intrinsic spin and polarization properties of a QD in a NW. We introduce a theoretical model which intuitively explains our experimental findings and shows how polarization is affected by various parameters such as NW diameter, dielectric constant of the surroundings and photon wavelength. As an example, we demonstrate access to the spin properties of a Zeeman split exciton in a QD by measuring the right- and left-hand circular photon polarization.

For our experiments we used single  $\text{InAs}_{0.25}\text{P}_{0.75}$  QDs embedded in InP NWs, grown by means of metal-organic vapor-phase epitaxy [25, 26, 27, 28]. Colloidal gold particles of 20 nm diameter were deposited on a (111)B InP substrate as catalysts for vertical NW growth. The diameter of the NW and the QD was controlled by the gold particle size, while the NW density was set by the gold particle density on the substrate. The QD height and NW length were determined by growth time [8]. By controlling diameter, height, and As concentration one can tune the QD emission in the wide range of 900 nm to 1.5  $\mu\text{m}$  [9]. Under appropriate growth conditions we were able to grow a sample with low density of NWs with a single QD that emits around 950 nm. In Fig. 1a) we show a scanning electron microscope (SEM) image of an as-grown sample with a low density of NWs, enabling optical study of a single NW QD. The NW length is about 2  $\mu\text{m}$ . A transmission electron microscope (TEM) image of a NW QD is shown in Fig. 1b). The QD is typically 9 nm high with a diameter of 31 nm, determined by energy dispersive X-ray analysis in a TEM. To simplify TEM studies, the InP section following QD growth was reduced, while for the samples used in our experiments, the QD was centered in the NW.

As-grown samples with vertically oriented NWs, referred as standing NWs, were used for experiments where the excitation (emission) was directed (measured) along the NW. We also transferred NWs to a Si substrate with a 290 nm  $\text{SiO}_2$  top layer, referred as lying NWs, for the experiments where light was directed perpendicular to the NW. Both samples were studied under the same experimental conditions. Micro photoluminescence (PL) studies were performed at 4.2 K. The NW QDs were excited

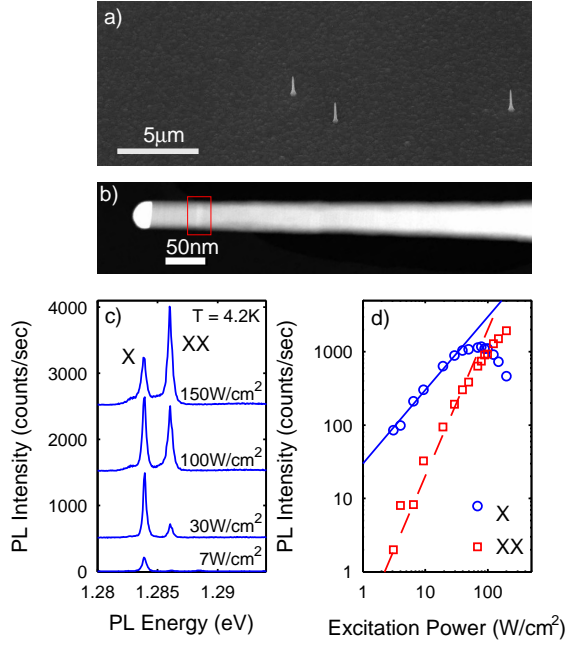


FIG. 1: (color online) a) SEM image of an as-grown sample with low density of standing NW QDs. b) TEM image of a NW QD. The QD is indicated by the red rectangle. The low contrast of the QD is due to the low As concentration. c) PL spectra of a single standing NW QD for various excitation powers. The two emission lines denoted with X and XX correspond to the exciton and biexciton emission, respectively. d) PL intensity of X and XX transitions as a function of the excitation power. The solid (dashed) line is a guide to the eye for linear (quadratic) power dependence.

with a linearly polarized 532 nm cw laser focused to a spot size of 1 μm using a microscope objective with a numerical aperture NA=0.85. The PL signal was collected by the same objective and was sent to a spectrometer, which dispersed the PL onto a nitrogen-cooled silicon array detector, enabling 50 μeV resolution. Linear and circular polarizations were determined with a fixed polarizer together with a half- and quarter-waveplate, respectively.

In Fig. 1c) we show PL spectra of a single standing NW QD for various excitation powers. At low excitation power only one emission line is visible at 1.284 eV, denoted as X. With increasing excitation power a second emission line, denoted as XX, emerges 2.2 meV above X. The PL intensities of X and XX as a function of excitation power, represented in Fig. 1d), show that X (XX) increases linearly (quadratically) with excitation power and saturates at high excitation powers. This behavior is typical for the exciton and biexciton under cw excitation. The spectral linewidths for various NW QDs in our samples are in the range of 50-200 μeV, demonstrating good quality of the NW QDs. Similar spectral characteristics are also found for lying NW QDs.

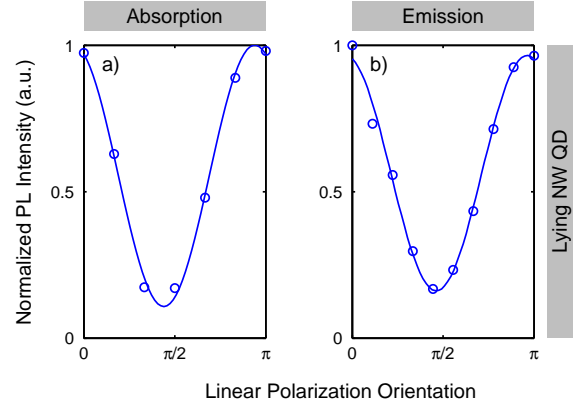


FIG. 2: (color online) Integrated PL intensity of the exciton in a lying NW QD a) as a function of the orientation of the linear excitation polarization and b) by probing the orientation of the linear emission polarization. Zero angle of orientation was chosen to match the NW elongation, determined by imaging. The solid curves are  $\sin^2$  fits used to extract the degree of linear polarization from minima and maxima of the fit.

In Fig. 2 we show the integrated PL intensity of the exciton in a lying NW QD as a function of linear excitation and emission polarization orientation. In Fig. 2b) the excitation polarization is set parallel to the NW elongation. However, the emission polarization is independent of the excitation polarization. We fit this data to a  $\sin^2$ -function with amplitude, offset, and phase as fit parameters. The degree of linear polarization,  $\rho = \frac{I_{max} - I_{min}}{I_{max} + I_{min}}$ , is obtained from the maxima and minima of the fit. Clearly, lying NW QDs show a large degree of linear polarization parallel to the NW, for both absorption and emission.

Polarization-sensitive PL measurements were performed on a number of NW QDs, both lying and standing, which are shown in Fig. 3a). The upper two graphs show the statistics of absorption and emission for lying NW QDs, for which absorbed (emitted) light is directed (measured) perpendicular to the NW elongation. The average degree of linear polarization is  $\rho_{abs} = 0.78 \pm 0.06$  ( $\rho_{emi} = 0.72 \pm 0.08$ ) for absorbed (emitted) light.

The same measurements were performed on standing NW QDs, for which light absorbed (emitted) by the QD is directed (measured) parallel to the NW elongation. The results are shown in the lower two graphs of Fig. 3a) and give a degree of linear polarization for absorbed (emitted) light of  $\rho_{abs} = 0.1$  ( $\rho_{emi} = 0.2$ ). This low value is expected for the symmetric structure of the NWs for light incident or emitted in the direction along the NW elongation. In addition, the measured degree of circular polarization of the QD emission is also low, 0.1.

To explain our experimental results for absorption by a lying NW QD, we use Mie theory for light scattering on dielectrics of cylindrical shape [29, 30]. The NW is modeled as an infinite cylinder. This approximation is

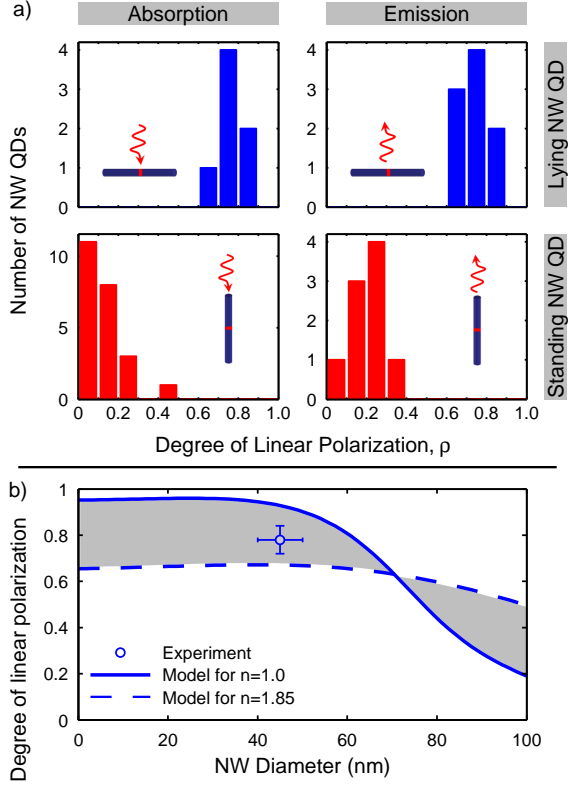


FIG. 3: (color online) a) Statistics of the degree of linear polarization for lying and standing NW QDs. The lying NW QDs (shown in blue) show high degree of linear polarization, about 0.8 and 0.7, for absorbed and emitted light, respectively. In contrast, the degree of linear polarization is low for standing NW QDs (shown in red), about 0.1 and 0.2 for absorbed and emitted light, respectively. Insets illustrate the NW orientations with respect to the light direction. b) Calculated degree of linear polarization in absorption as a function of NW diameter. In this case incident light is directed perpendicular to the NW elongation, as for the lying NW QDs. For the solid (dashed) curve an effective refractive index of  $n = 1$  ( $n = 1.85$ ) is used.

valid as long as the nanowire diameter is much smaller than its length, as in our case. We calculate the scattering and absorption of light using the scalar wave equation in cylindrical coordinates. We consider two cases: incident electric field parallel or perpendicular to the NW elongation. We extract the scattered field for both polarizations from the wave equation, using the boundary conditions at the interface of two different dielectrics [31], i.e., at the NW surface. From this field, we calculate for both cases the cross-sections  $Q_{sca}$  and  $Q_{ext}$  for scattering and extinction. The absorption cross section is obtained by  $Q_{abs} = Q_{ext} - Q_{sca}$  for both polarizations. To take into account the illumination of the NW through a microscope objective, we integrate the average absorption cross section over angles of incidence with respect to the

NW elongation comprised between  $\arccos(\text{NA})$  and  $\pi/2$ .

Mie theory assumes the surrounding of the NW as a homogeneous medium, which differs from our situation where the NW is lying on a substrate. Therefore, to approximate the effect of the substrate we consider the nanowire as being embedded in a medium with an effective refractive index, i.e., an average of the refractive indices of the different media surrounding the NW: vacuum,  $\text{SiO}_2$ , and Si. The outcome of the calculations, assuming an effective refractive index of  $n_{eff} = 1.85 = 0.5n_{vacuum} + 0.25n_{\text{SiO}_2} + 0.25n_{\text{Si}}$  is represented by the dashed curve in Fig. 3b). As an upper limit we consider the NW in vacuum, thus ignoring the substrate, which is represented by the solid curve in Fig. 3b). For the calculations we use our actual excitation wavelength of 532 nm. Our experimental value lies in between the two curves, having a surrounding refractive index consisting of a mixture of vacuum, Si, and  $\text{SiO}_2$ . As can be seen in Fig. 3b) one can increase the degree of linear polarization by measuring NW QD in vacuum, or decrease it by increasing the NW diameter. However, in the latter case the advantage of the one-dimensional channel of the device is reduced as well.

The NW geometry is not the only source of polarization anisotropy. Calculations by Niquet and Mojica [32] show that the polarization properties are strongly affected by the aspect ratio of the QD dimensions, due to strain originating from the lattice mismatch between the NW and the QD. However, in our case the strain is negligible due to the low phosphorus content and the main contribution to polarization anisotropy comes from the NW geometry.

The low degree of linear polarization for vertical NW QDs implies that the NW does not affect the polarization of photons radiated from the QD, thus allowing access to intrinsic spin and polarization properties of NW QDs. To experimentally demonstrate this we measure the polarization of photons emitted from a Zeeman split exciton in a NW QD. In Fig. 4b) we show PL of the NW QD exciton under external magnetic field  $B$  in the Faraday configuration ( $B \parallel \text{NW}$ ). Clearly, the exciton undergoes a Zeeman splitting and diamagnetic shift. From these measurements we extract the exciton  $g$ -factor,  $g = 1.3 \pm 0.1$ , which is typical for our sample. Polarization properties of the split exciton lines at 9 Tesla are shown in Fig. 4a). The high energy emission line is right-hand circularly polarized, while the lower energy emission line is left-hand circularly polarized. These two polarization states,  $\sigma^+$  and  $\sigma^-$ , correspond to the two different spin states of the exciton  $\downarrow\uparrow$  and  $\uparrow\downarrow$ , respectively, where  $\uparrow$  ( $\uparrow$ ) represents a spin-up electron (hole) and  $\downarrow$  ( $\downarrow$ ) represents a spin-down electron (hole). These results show that the emission polarization of a QD is not obscured by the NW geometry, and that electron-hole spin states in a QD are properly converted to the polarization states of the emitted photons. In contrast, measurements on a lying NW

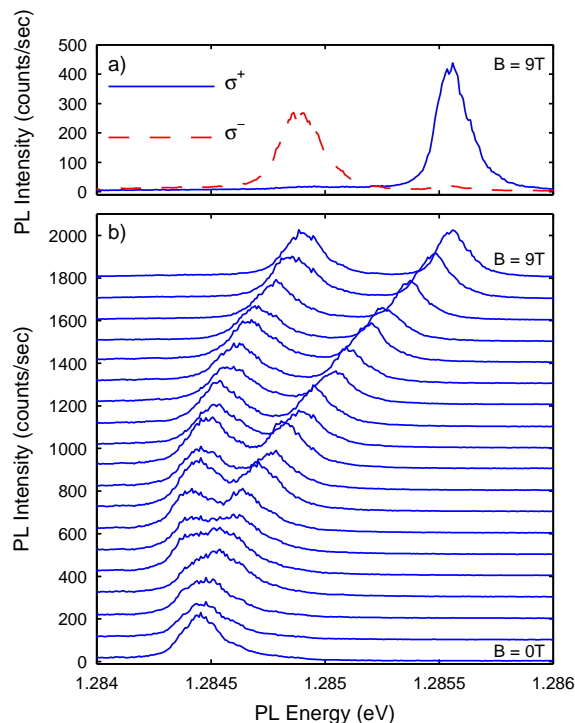


FIG. 4: (color online) a) Polarization sensitive PL of a standing NW QD at 9 T. The solid (dashed) curve represent right- (left-) hand circularly polarized exciton emission, denoted by  $\sigma^+$  ( $\sigma^-$ ). b) PL of a standing NW QD under external magnetic field. Magnetic field is varied between 0 and 9 T in steps of 0.5 T.

QD show that the two emission lines of a Zeeman split exciton are strongly linearly polarized, with respect to the NW elongation, as expected (not shown).

To conclude, we have correlated the polarization of light absorbed and emitted by a QD embedded in a NW with its propagation direction with respect to the NW elongation. We found that the polarization of the absorbed (emitted) light, when directed (measured) perpendicular to the NW elongation, is affected by the NW geometry and is strongly linearly polarized along the NW. In contrast, this is not the case for the configuration where the absorbed (emitted) light is directed (measured) parallel to the NW elongation. In this configuration the intrinsic spin and polarization of the NW QD can be accessed. We have demonstrated this by measuring polarization states of photons radiated from a Zeeman split exciton in a NW QD.

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